PB90-143017

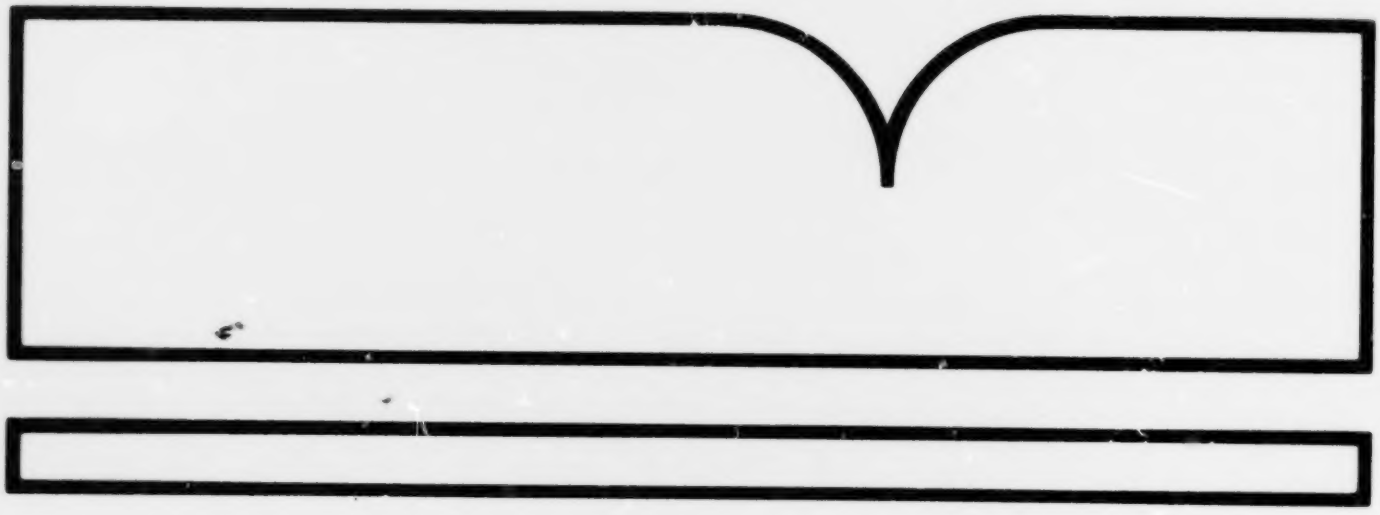
Relationship between Mesoscale Acid Precipitation and Meteorological Factors

Wisconsin Univ.-Madison

Prepared for:

Environmental Research Lab.-Duluth, MN

1982



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TECHNICAL REPORT DATA
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1. REPORT NO.	2.	PB90-143017
EPA/600/D-89/257		

4. TITLE AND SUBTITLE	5. REPORT DATE
Relationship Between Mesoscale Acid Precipitation and Meteorological Factors	5. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
Ragland, K.W. and K.E. Wilkening	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Department of Mechanical Engineering University of Wisconsin Madison, WI 53706	11. CONTRACT/GRANT NO. CR 809412

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Environmental Research Laboratory Office of Research and Development U.S. Environmental Protection Agency Duluth, MN 55804	14. SPONSORING AGENCY CODE EPA-600/03

15. SUPPLEMENTARY NOTES

Published in: Acid Precipitation: Effects of Ecological Systems, Chapter 6, F.M. D'Itri (Ed.)., Ann Arbor Science, Ann Arbor, MI, pp. 123-139 (1982).

16. ABSTRACT

This chapter deals with sulfur dioxide and sulfate deposition from the atmosphere to the ground. Sulfur dioxide may be transformed to sulfate compounds in the atmosphere and in vegetation, soil, and water. The extent to which sulfur deposition changes the pH of the soil and water is not addressed here. Rather, sulfur loading is taken as an indicator of potential for acidification. Nitrogen compounds may also produce acidification; however, less is known about these processes and they are not considered here.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to public	Unclassified	21
	20. SECURITY CLASS (This page)	22. PRICE
	Unclassified	

EPA Form 2220-1 (Rev. 4-77)

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EPA/600/D-89/257

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ACID PRECIPITATION

Effects on Ecological Systems

Edited by
FRANK M. D'ITRI

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230 Collingwood, P.O. Box 1425, Ann Arbor, Michigan 48106

Library of Congress Catalog Card Number 81-69072
ISBN 0-250-40509-1

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Kent TN15 8PH, England

CHAPTER 6

RELATIONSHIP BETWEEN MESOSCALE ACID PRECIPITATION AND METEOROLOGICAL FACTORS

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INTRODUCTION

This chapter deals with sulfur dioxide and sulfate deposition from the atmosphere to the ground. Sulfur dioxide may be transformed to sulfate compounds in the atmosphere and in vegetation, soil, and water. The extent to which sulfur deposition changes the pH of the soil and water is not addressed here. Rather, sulfur loading is taken as an indicator of potential for acidification. Nitrogen compounds may also produce acidification; however, less is known about these processes and they are not considered here.

Acid precipitation can occur when particles and gases are removed from the atmosphere (1) by rain and snow (wet deposition), and (2) by impaction on water, soil, and vegetation surfaces (dry deposition). The transformation of gases to particles in the atmosphere and the deposition rates of gases and particles are a function of meteorological conditions over a period of days as well as the distribution of emission sources.

To justify controls on emissions of acid precursors, the relationship between a particular source and the deposition pattern that it produces needs to be understood. This is difficult to accomplish by field monitoring because monitoring data register many sources, some of which may be hundreds or thousands of kilometers away. Mathematical models are the only way to estimate the relative contribution of different sources to the total deposition at a particular receptor.

Atmospheric deposition models may be divided into (1) long-range transport models, where the region of interest extends

over thousands of kilometers, say the eastern half of the U.S. and Canada, and (2) mesoscale models where the analysis region extends over hundreds of kilometers, say the state of Michigan. The focus here is on mesoscale models. Because mesoscale models can provide higher resolution and more detailed simulation of meteorological conditions, they can predict the influence of a specific emission source on deposition at a nearby receptor point. However, mesoscale models show only incremental changes within the region rather than absolute deposition rates.

The purpose of this chapter is to briefly describe our mesoscale deposition model (UWATM-SOX) and present the results of the application of this model to the Rainy Lake watershed, which is located northwest of Lake Superior. Annual, seasonal, and weekly calculations of sulfate deposition are presented and compared with field monitoring data.

DESCRIPTION OF THE MODEL

The UWATM-SOX model is a time-dependent, cell-type model which numerically solves coupled sulfur dioxide and sulfate ion diffusion in an Eulerian (fixed) frame of reference in the atmospheric boundary layer. Included are the processes of advection, turbulent diffusion, chemical reactions, source input, and dry and wet deposition (Figure 1). The numerical solution is a first-order fully implicit finite difference scheme.

The model is developed for a mesoscale (10s to 100s of kilometers) analysis. The region of study is divided into a uniform horizontal two-dimensional grid network. The rectangular grid size and number depends on the extent of the region, the desired cost of the computer runs, and the desired accuracy of analysis. Above the grids are six vertical cells which extend from ground level to an upper limit called the mixing height. The model uses three lower fixed-height (50 m) and three upper variable-height cells. Therefore, the space enclosing the region of study is completely partitioned by a three-dimensional cell structure. The single cell is the basic unit within which the model calculations are performed.

The model inputs emissions once at the beginning of the simulation and inputs meteorological data every hour. Calculations are performed every twenty minutes within an hour for time periods up to one year. Each emission source is assigned to one of the grids. A plume rise is calculated and the resulting "effective stack height" is placed within the appropriate vertical cell. The meteorological condition, known

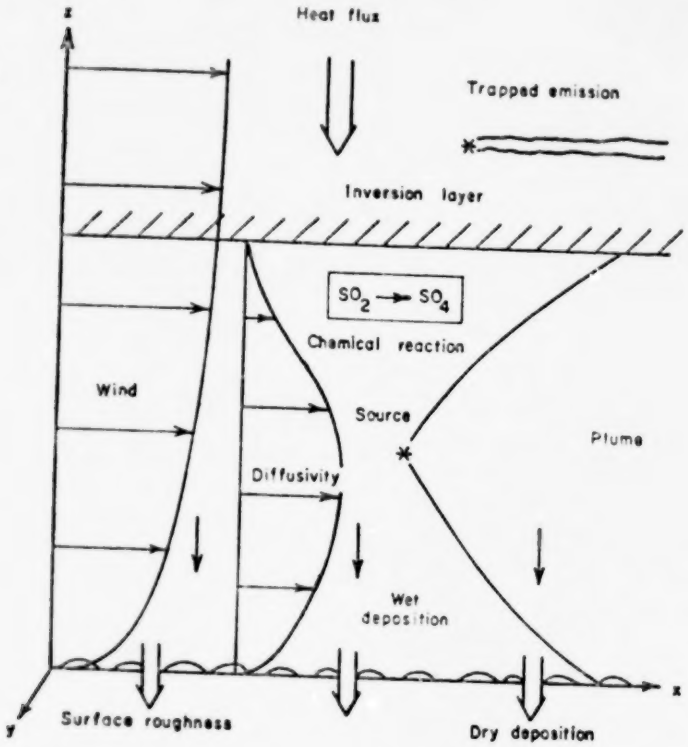


Figure 1. Schematic diagram of atmospheric mechanisms.

as lofting (effective stack height above mixing height) is accounted for.

The meteorological data are obtained on tapes from the National Climatic Center (NCC) in Asheville, North Carolina. The tapes required are: hourly surface observations, twice daily mixing heights, and hourly precipitation. These data are consolidated and pre-processed by three tape-handling programs and a program (METDATA) which converts the data to the hourly format required by the model.

The atmospheric boundary layer is characterized by a set of wind speed and eddy diffusivity profile equations which are calculated from ground level to the top of the mixing layer (Ragland et al., 1977). The profiles describe the transport and

diffusion behavior of the pollutants and are assumed to be vertically variable but horizontally uniform across the region of study over a 1-hr period. The profiles are dependent on the wind speed and stability of the atmosphere. The boundary layer is subdivided into a surface layer and a second layer extending up to the mixing height. The surface layer equations are based on the Monin-Obukov similarity theory. In the upper layer, the wind profile is a form of the power-law and the diffusivity is assumed constant. The profiles are calculated from the geostrophic wind speed, net heat flux to the ground and surface roughness. The geostrophic wind is calculated from the surface wind speed using a modified Regula Falsi iteration scheme. The net heat flux is calculated by first using the NCC surface observations data and the Pasquill-Turner method of stability classification to determine the stability class, and then assigning a heat flux value to each class. A single surface roughness value is chosen to characterize the region.

The chemical conversion from sulfur dioxide to sulfate ion is assumed to be first-order. At present, separate day and night reaction rates are used although hourly and seasonal changes are possible if data are available.

The dry deposition is handled by means of a deposition velocity which is calculated as a function of both terrain and meteorological conditions. Deposition velocity changes hourly, but is uniform across the region. It is determined from an aerodynamic resistance which depends on meteorological conditions close to the ground, and a surface resistance which depends on pollutant species and the nature of the surface (snow, water, vegetation, etc.). Surface resistances are input by the user and aerodynamic resistance is internally calculated from wind profile information.

The wet deposition scheme assumes all precipitation forms according to the Bergeron or cold cloud process. Ice crystals form in the upper portion of the cloud, and as they sweep downward through the cloud, they capture cloud droplets containing sulfate and dissolved sulfur dioxide. The sulfur eventually incorporated into the precipitation is drawn in through the cloud base. Three equations for the wet deposition removal rate are used; one for rain or snow removal of sulfate, and one each for rain and snow removal of sulfur dioxide (Glass and Loucks, 1980). Precipitation is assumed to occur uniformly across the region. At present, there is no analysis for fractional area covered by a precipitation event.

There are several computer techniques worth noting. First, handle the numerical calculations for arbitrary wind

directions, a dual grid system is used, one fixed in position and one which overlaps the fixed grid and rotates with the wind angle. Transfers of concentrations are performed back and forth between the two systems as the wind direction shifts. This procedure minimizes so-called numerical diffusion (Lio, 1980). Second, the main program has a "stop-start" capability such that progress can be stopped anywhere in the stream of calculation and started exactly where it left off with no outside intervention other than restarting the run. Third, extensive use has been made of common blocks and the Fortran procedure (PROC) statement to eliminate unnecessary interprogram storage locating calls and to facilitate internal changes which affect many programs at once. Fourth, a very complete record of the values of all significant variables is stored on tape hourly for future analysis. The stored information includes the date, ground level concentration and deposition arrays, and certain meteorological, emissions, and regional mass balance data.

The model results include 3-hr, 24-hr, seasonal and annual average ground level concentrations, and dry, wet, and total depositions of sulfur dioxide and sulfate. The results are printed grid by grid in an array format and isopleths are computer-plotted. The results also include a frequency distribution of concentration/deposition values for any selected grid point. Finally, a regional atmospheric sulfur budget is computed which analyzes, by total mass and percentage of sulfur input, the flow of sulfur (SO_2 and SO_4^{2-}) into, out of, and within the region.

The model is written in ASCII Fortran and has been run on the Madison Academic Computing Center's SPERRY UNIVAC 1100/82 series computer. In an application of the model in which a cell structure of 11 x 13 x 6 was used with hourly changes in meteorological data and a 20-min time step, the main program required 50K core space, and a 1-yr simulation took 6 hr of CPU time.

An overview of the model is shown schematically in Figure 2. A summary of the user-supplied model input parameters and the values which we have used is given in Table 1.

CASE STUDY - THE RAINY LAKE WATERSHED

The Rainy Lake watershed is located in northeastern Minnesota and western Ontario, and includes the Quetico Provincial Park, the Boundary Waters Canoe Area (BWCA), and the Voyagers National Park. This area lies within 2 days travel of 50 million people and yet contains extensive prime wilderness

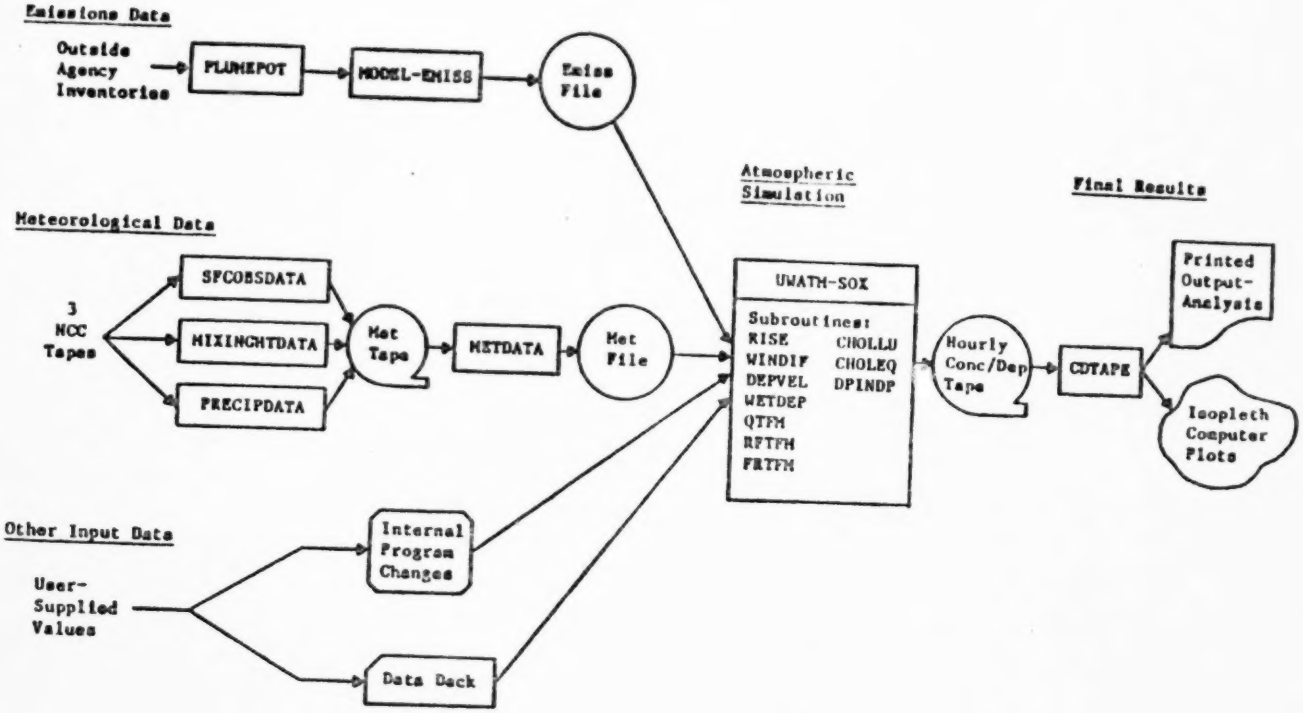


Figure 2. Schematic of the modeling operation.

Table 1

Summary of Model Parameters Used

Parameter	Value
SO_2 chemical transformation rate (%/hr):	Night 0.5, day 2.0
SO_2 dry deposition surface resistance (sec/cm):	Summer 0.5, winter 3.0
SO_4^{2-} dry deposition surface resistance (sec/cm):	Summer 0.5, winter 5.0
SO_2 rain deposition rate (%/hr): where P = rain rate (mm/hr)	$33.0\ P^{0.90}$
SO_2 snow deposition rate (%/hr):	$16.5\ P^{0.90}$
SO_4^{2-} rain and snow deposition rate (%/hr):	$33.0\ P^{0.63}$
Mixing height (m):	Hourly values calculated from twice daily soundings.
Wind data (m/sec):	Six levels calculated hourly from wind speed, direction, temperature, and cloud cover surface observations.
Diffusivity data (m^2/sec):	Six levels calculated hourly from wind speed, temperature, and cloud cover surface observations for vertical and laterial diffusivity.
Cell size (km):	30 x 30
Number of cells:	11 x 13

areas. The BWCA and adjoining areas are believed to be sensitive to acid precipitation (Glass and Loucks, 1980).

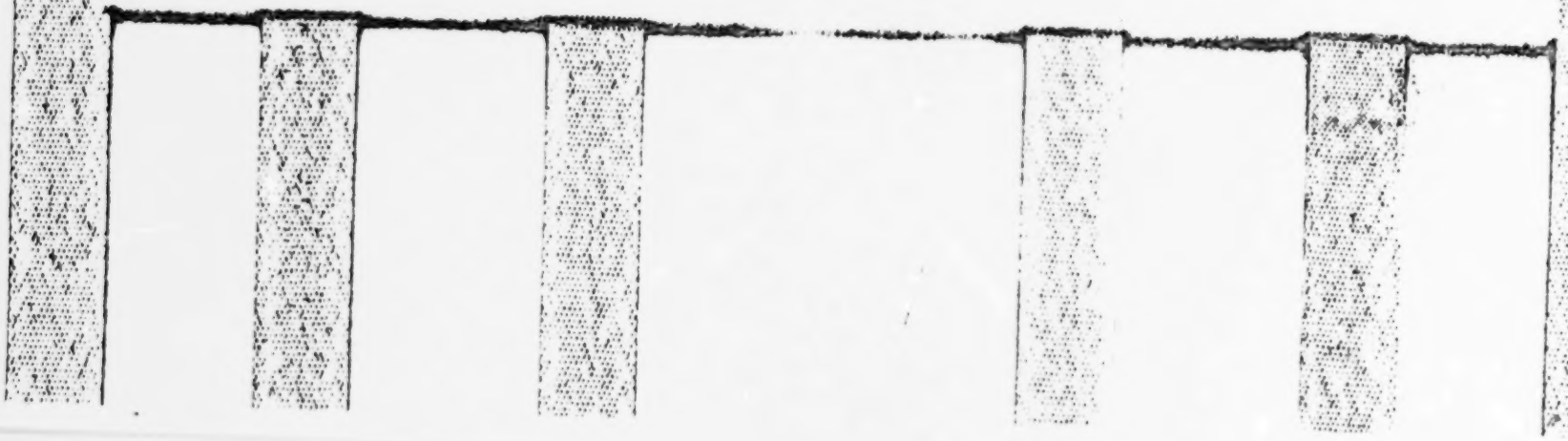
The modeling area we have chosen is the 330 x 390 km region shown in Figure 3. The Rainy Lake watershed is shown in dotted lines and major emission sources in Duluth, the Missabe Iron Range, International Falls, and Atikokan are indicated. The sulfur dioxide emissions inventory for the region is given in Table 2 (Price, 1980). The total emissions of sulfur dioxide due to point sources in the region in 1976 were 3232 gm/sec or 102,000 tonne/yr. In 1978, the sulfur dioxide emissions were 3139 gm/sec or 99,000 tonne/yr. The area source emissions of

Table 2

Emission Inventory for the Study Region

Source	Location	Stack Height (m)	1976 Sulfur Dioxide Emissions (gm/sec)	1978 Sulfur Dioxide Emissions (gm/sec)
Portlach NW	Cloquet	91	34	0
Portlach NW	Cloquet	11	22	0
Continental Oil	Cloquet	27	20	27
Continental Oil	Cloquet	41	21	22
Conved	Cloquet	22	8	0
Erie Mining	Taconite Harbor	107	3	0
Minn. P&L	Clay Boswell	67	542	400
Boise Cascade	Intl. Falls	213	939	965
Two Harbors W&L	Two Harbors	61	26	11
Reserve Mining	Silver Bay	46	101	0
Erie Mining	Hoyt Lakes	66	101	74
Minn. P&L	Aurora	40	35	24
Minn. P&L	Duluth	91	192	215
Minn. P&L	Duluth	101	20	67
Duluth Steam	Duluth	66	29	0
Hibbing-Public UT	Hibbing	72	23	14
Virginia Public UT	Virginia	12	10	42
Superwood	Duluth	40	54	43
U.S. Air Force	Duluth	24	7	6
Reserve Mining	Babbit	9	4	0
		17	3	0

U.S. Steel Coke	Duluth	69	109	123
U. of Minn.	Duluth	18	3	0
U.S. Steel Shipping	Duluth	8	9	9
Ontario-Minn. P&L	Fort Francis	76	26	27
Steep Rk.-Caland Ore	Atikokan	85	861	863
Lake Superior P	Ashland	43	45	80
Murphy Oil	Superior	63	58	41
Superior W, L&P	Superior	46	13	14
U. of Wisconsin	Superior	69	9	0
CLM Corp.	Superior	20	9	24
Hanna Minn.	Hibbing	37	0	3
U.S. Steel	Mt. Iron	51	0	5
Eveleth Tac.	Eveleth	47	0	6
Hibbing Tac.	Hibbing	39	0	20
Inland Steel	Virginia	41	0	10
American Can	Ashland	22	0	4
TOTAL			3232	3139



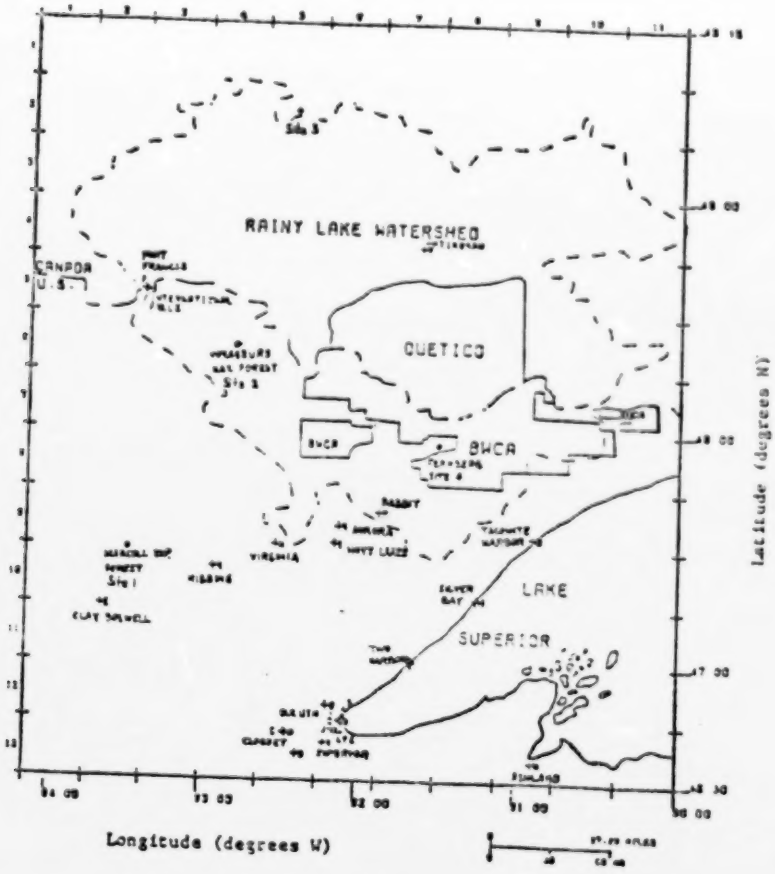


Figure 3. Map of the study region.

sulfur dioxide due to space heating and small industrial and commercial sources were not available, but are believed to be small in this region. Direct sulfate emissions were assumed to be 3 percent of the sulfur dioxide emissions by weight, which is a generally observed result from stack sampling.

The regional sulfur dioxide emissions may be compared to the Minnesota state-wide total of 521,000 tonne/yr, Wisconsin 937,000 tonne/yr, and Illinois 2,344,000 tonne/yr. The question to be answered is how much of the regional sulfur emissions are

deposited within the region and how does this compare to the monitoring data on an annual, seasonal, and weekly basis.

Before discussing the deposition results, the ambient air concentrations will be briefly mentioned. The annual average sulfur dioxide concentrations away from any sources were calculated to be about 0.5 $\mu g/m^3$, while ambient sulfate concentrations were about 10 times smaller. The 24-hr worst-case sulfur dioxide concentrations away from any sources were about 5 $\mu g/m^3$ and the 24-hr worst-case sulfate concentrations were also about 5 $\mu g/m^3$

The annual (1976) wet deposition isopleths for the region are shown in Figure 4 and the annual wet plus dry deposition isopleths are shown in Figure 5. To express deposition as kilograms of sulfate per hectare, the sulfur dioxide deposition was multiplied by 1.5 (the ratio of molecular weights) and combined with the sulfate values. The regional mass balance, presented in Table 3 for 1976, shows that 17 percent of the sulfur emissions was deposited in the region due to dry deposition and 1.3 percent was retained due to wet deposition, whereas 82 percent of the sulfur emissions was transported out of the region annually. Of the sulfur deposited in the region, 8.5×10^9 gm out of 9.4×10^9 gm (77%) were deposited by dry deposition of sulfur dioxide.

In this region, rain occurs from May through October, while snow occurs from November through April. During 1976, there was 27.2 cm of rain compared with a normal of 40 cm, and there

Table 3.

Regional Mass Balance for 1976

Origin	Sulfur Dioxide	Sulfate	Sulfur
	----------10^9 gm/yr----------		
Emissions	102	3.1	52.3
Chemical reaction	6	9.8	-
Dry deposition	17	0.6	8.7
Wet deposition	1	0.5	0.7
Transport out of region	78	11.8	42.9

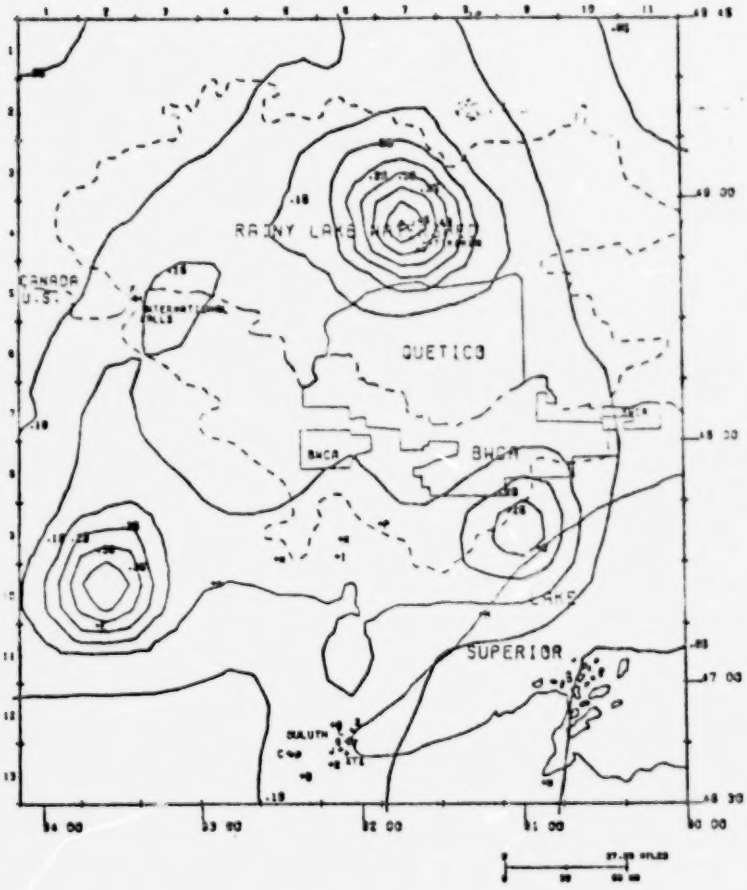


Figure 4. Computed annual wet deposition of sulfate and sulfur dioxide (kg SO_4^{2-}/ha).

was 9.0 cm of snow compared with a normal of 15 cm. Hence, there was 34 percent less precipitation than normal during 1976.

It is of interest to compare the deposition during the snow season (November to April) with the rain season (May to October). The calculated total deposition (wet plus dry) of sulfur dioxide and sulfate plotted as sulfate is shown in Figure

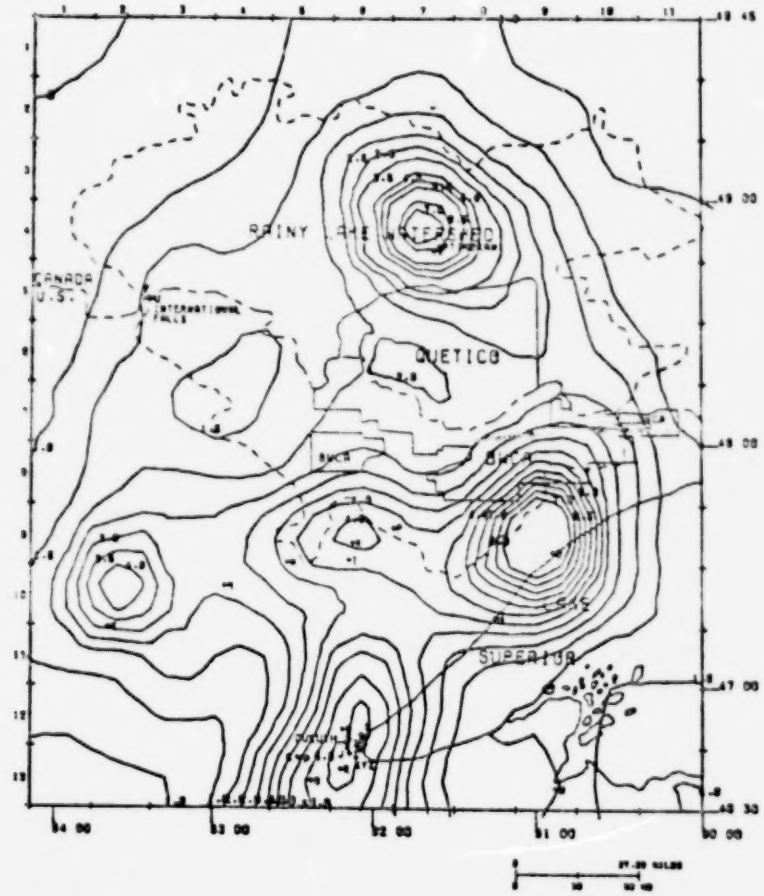


Figure 5. Computed annual wet plus dry deposition of sulfate and sulfur dioxide (kg SO_4^{2-}/ha).

5. The calculations are summarized for grid (2,9) the Marcell, Minnesota, site in Table 4. The greatest percentage of sulfur deposition occurs by dry deposition of sulfur dioxide during the summer. Wet deposition of sulfate was comparable in winter and summer, although total wet deposition of sulfate and sulfur dioxide was greater in the summer. It is believed that the reason wet deposition of sulfate is similar in winter and summer

Table 4

Calculated Regional Deposition at the Marcell, Minnesota, Site and Comparison with Measurements

	Dry	Wet	Wet & Dry
	----------kg/ha----------		
UWATM-SOX MODEL, 1976			
Winter SO_2 deposition	0.24	0.01	0.25
Summer SO_2 deposition	1.23	0.10	1.33
Winter SO_4 deposition	0.02	0.03	0.05
Summer SO_4 deposition	0.02	0.03	0.05
Winter deposition as SO_4	0.36	0.05	0.41
Summer deposition as SO_4	1.86	0.18	2.04
Annual deposition as SO_4	2.22	0.23	245
MEASUREMENTS			
Annual deposition as SO_4	-	15.6[a] (11.5[b])	-
Snow deposition as SO_4	-	1.6[a] (2.2[b])	1-2[c]
EASTERN NORTH AMERICA MODELS[d]			
Annual deposition as SO_4	1.8-7.2	1.9-4.5	3.7-11.7

[a]National Atmos. Deposition Program data July 1979 through June 1980.
[b]NADP data April 1978 through March 1978.
[c]Snow cores (65) in BWCA, 1978-1978 (Glass, 1979).
[d]Preliminary report (Ferguson and Machta, 1981).

is that there are more hours of precipitation in winter than in summer although the total amount of precipitation is greater in the summer.

The computer calculations of sulfate deposition are compared with measured deposition for the Marcell, Minnesota, site (Table 4). Unfortunately, the computer calculations for the time period of the measurements (1978 to 1980) have not been completed so that calculations for 1976 will have to be used. The precipitation for 1978 was 96 percent of normal.

The National Atmospheric Deposition Program (NADP) has a site in the region at Marcell, Minnesota, starting in July 1978. The NADP site provides wet sulfate deposition on a weekly basis. The total wet sulfate deposition at the Marcell site was calculated to be 0.23 kg/ha, whereas the monitoring value was 15.6 kg/ha for 1979 to 1980 and 11.5 kg/ha for 1978 to 1979. The main difference is due to transport of sulfate from outside the region. During the snow season, the calculated wet sulfate deposition was 0.05 kg/ha, compared to measured values of 1.6 and 2.2 kg/ha. Again, the measured value represented interregional and intraregional transport.

Snow core samples taken at the end of the snow season provide valuable data on wet plus dry deposition of sulfate. Sixty-five snow cores taken in April, 1979, in the Boundary Waters Canoe Area were mostly in the range of 1 to 2 kg/ha sulfate, which may be compared to the calculated value of 0.5 kg/ha (Glass, 1979).

Finally, Table 4 presents deposition calculations from long-distance transport models of eastern North America which show a range of 1.9 to 4.5 kg/ha for wet deposition at the Marcell site and 3.7 to 11.7 kg/ha for wet plus dry deposition (Ferguson and Machta,1980). Our regional model gave 2.4 kg/ha for the total annual sulfate deposition.

The wet and dry deposition fluxes may also be compared on a weekly basis. In Table 5, the regional mass balance is summarized for the extreme or worst-case conditions of a clear summer week, a wet summer week, a clear winter, and a winter week with snowfall during 1976. The rainy week had 7 hr of rainfall, producing 4.3 cm of water. The snowing week had 32 hr of snowfall, producing 1.7 cm of water. The dry deposition is three times greater in summer than in winter, whereas the surface resistance for sulfur dioxide was six times less in summer than in winter (Table 1). The wet deposition is four and one-half times greater in summer than in winter. During the wet summer week, 65 percent of the sulfur emissions is transported out of the region, while during a winter week, more than 90 percent is transported out of the region.

During the wet summer week (7/19/76 to 7/25/76), the calculated wet deposition flux of sulfur dioxide and sulfate as sulfate was 0.06 kg/ha at the Marcell site. By way of comparison, a wet deposition flux of 2.2 kg SO_4^{2-}/ha was recorded during a week in July of 1979 at the Marcell site. Since the total measured for the year was 11.5 kg SO_4^{2-}/ha at Marcell, it is clear that a few big events coming from extraregional buildup of sulfate produce much of the wet deposition loadings.

Table 5

Mass Balance for Worst-Case Weeks

	Dry Deposition	Wet Deposition	Transport Out of Region
	----------10^8 gm S----------		
Clear summer week	3.0	0	7.1
Wet summer week	2.6	0.9	6.6
Clear winter week	1.0	0	9.1
Snowing winter week	0.7	0.2	9.2

ACKNOWLEDGMENT

This work was supported by a grant from the U.S. Environmental Protection Agency, Environmental Research Laboratory, Duluth, Minnesota, under the direction of Dr. Gary E. Glass.

LITERATURE CITED

Ferguson, H.L. and L. Machta. 1980. Preliminary Report of Atmospheric Modeling Work Group 2. U.S. Environmental Protection Agency, Washington, D.C. 20460.

Glass, G.E. 1979. Unpublished Data. U.S. Environmental Protection Agency, Environmental Research Laboratory, Duluth, Minnesota 55804.

Glass, G.E. and O.L. Loucks (eds). 1980. Impacts of Airborne Pollutants on Wilderness Areas Along the Minnesota-Ontario Border. EPA-600/3-80-044, U.S. Environmental Protection Agency, Environmental Research Laboratory, Duluth, Minnesota 55804.

Lio, P.I. 1980. A Two-Dimensional Sulfur Dioxide/Sulfate Air Quality Model and Comparison With a Three-Dimensional Model. M.S. Thesis, University of Wisconsin, Madison, Wisconsin 53706. 104 pp.

Price, B. 1980. Personal Communication, Minnesota Pollution Control Authority Emission Inventory Data. Minnesota Pollution Control Authority, Minneapolis, Minnesota.

Hegland, K.W., R.L. Dennis, and K.E. Wilkening. 1977. Boundary Layer Model for Transport of Urban Air Pollutants. AIChE Symposium Series 165, 73:1-10.